



RECEIVED
DEC 09 2003
TC 1700

DECLARATION

I, Shigeaki Nishikawa of Nishikawa & Associates, Visual City, Suite 401, 43-9, Higashi-Nippori 3-chome, Arakawa-ku, Tokyo, Japan, do solemnly and sincerely declare that I am well acquainted with both the Japanese language and the English language and that the attached English translation of an officially certified copy of Patent Application No. 142555/1999 is a true and correct translation to the best of my knowledge and belief from the Japanese language to the English language.

Dated this 2nd day of December, 2003

Shigeaki Nishikawa

(translator)

(Translation)

PATENT OFFICE
JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of
the following application as filed with this Office.

Date of Application: May 24, 1999

Application Number: Patent Application No. 142555/1999

Applicant(s): NIPPON ZEON CO., LTD.

Dated: June 2, 2000

Takahiko Kondo
Commissioner, Patent Office

Certificate No. 2000-3040955

[Document Name] Application for Patent
[Docket Number] PN990039
[To] Hon. Director General of Patent
Office
5 [International Patent Classification] B29C
[Inventor]
[Address or Domicile] c/o Research & Development
Center, Nippon Zeon Co., Ltd., 2-1, Yako 1-chome
Kawasaki-ku, Kawasaki-shi, Kanagawa
10 [Name] Naoki Shinohara
[Inventor]
[Address or Domicile] c/o Research & Development
Center, Nippon Zeon Co., Ltd., 2-1, Yako 1-chome
Kawasaki-ku Kawasaki-shi, Kanagawa
15 [Name] Keiichi Kawata
[Applicant for Patent]
[Identification Number] 000229117
[Name] NIPPON ZEON CO., LTD.
[Representative] Katsuhiko Nakano
20 [Indication of Fee]
[Deposit Account Number] 033684
[Fee] ¥21,000
[List of Materials Submitted]
[Material Name] Specification 1
25 [Material Name] Abstract 1

[Document Name] Specification

[Title of the Invention] LAMINATED FILM

[Claims]

5 [Claim 1] A laminated film comprising at least one layer containing a linear low density polyolefin having a long period of at most 275 angstroms as measured by the small angle X-ray scattering method, at least one alicyclic polymer layer and at least one thermoplastic resin layer.

10 [Claim 2] The laminated film according to Claim 1, wherein the linear low density polyolefin has a long period of at most 275 angstroms as measured by the small angle X-ray scattering method, and a lamellar thickness of at most 145 angstroms.

15 [Claim 3] A laminated film comprising at least one layer containing a linear low density polyolefin obtained by polymerization making use of a metallocene catalyst, at least one alicyclic polymer layer and at least one thermoplastic resin layer.

20 [Claim 4] A laminated film comprising at least one layer of a composition containing a linear low density polyolefin having a long period of at most 275 angstroms as measured by the small angle X-ray scattering method and an alicyclic polymer, at least one alicyclic polymer layer and at least one thermoplastic resin layer.

25 [Claim 5] A composition comprising a linear low density polyolefin having a long period of at most 275 angstroms as measured by the small angle X-ray scattering

method and an alicyclic polymer.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

5 The present invention relates to a laminated film,
and particularly to a laminated film free of clouding, high
in transparency and heat resistance, excellent in
tearability or cuttability, good in adhesion to others and
excellent in mechanical strength.

10 [0002]

[Prior Art]

Films made by vinyl chloride resins have heretofore
been used as films for food packaging, agricultural use,
drug packaging, etc. However, films containing materials
15 possibly produce substances such as dioxin and
environmental hormones, which exert an adverse influence on
life environment and living bodies, have been going to be
changed to resin films having high environmental safety
such as polyethylene, and many researches and developments
20 have come to be made.

In the course of such researches and developments,
there have been proposed single-layer films composed of a
alicyclic polymer such as a norbornene addition polymer and
laminated films obtained by laminating a polyethylene layer
25 and a norbornene addition polymer layer.

The single-layer films composed of the norbornene
addition polymer are high in transparency and low in

hygroscopicity. Since such a single-layer film is insufficient in oil resistance or resistance to greases and oils, however, the film tends to cause cracking and clouding when it is used in such a form that it comes into direct contact with food.

[0003]

On the other hand, a laminated films obtained by laminating different kinds of resins tend to delaminate each layers or lower its transparency, etc. For example, a laminated film obtained by laminating a norbornene addition polymer and a layer composed of polyolefin or polyamide has been easy to cause such inconvenience that when it is applied to use applications, in which the laminated film is stretched, white wrinkles occur at a stretched portion, or one layer is broken due to delamination because the elongation percentages of the individual layer are different and the compatibility of the norbornene addition polymer with polyolefin or polyamide is insufficient.

[0004]

A film comprising at least one composition comprising a polyolefin and a norbornene addition polymer has been proposed, however, it is difficult to provide a film free of clouding while retaining the desired strength and high transparency because of the low compatibility of the polyolefin and the norbornene addition polymer.

[0005]

[Problems Sought for Solution by the Invention]

It is an object of the present invention to provide a laminated film which is free of clouding, high in transparency and heat resistance, excellent in tearability or cuttability and low moisture permeability, good in
5 adhesion to others and excellent in mechanical strength.

The present inventors have carried out an extensive investigation with a view toward achieving the above objects. As a result, it has been found that when at least one layer containing a linear low density polyolefin having
10 a long period of at most 275 angstroms as measured by a small angle X-ray scattering method, at least one alicyclic polymer layer and at least one thermoplastic resin layer are laminated, a laminated film can be provided as a film free of clouding, high in transparency and heat resistance,
15 excellent in tearability or cuttability and good in adhesion to others, hard to cause delamination, excellent in mechanical strength. The present invention has been led to completion on the basis of this finding.

[0006]

20 [Means for solution of the Problems]

According to the present invention, there is thus provided a laminated film, comprising at least one layer containing a linear low density polyolefin having a long period of at most 275 angstroms as measured by a small
25 angle X-ray scattering method, at least one alicyclic polymer layer and at least one thermoplastic resin layer. There is also provided a laminated film, comprising at

least one layer containing a linear low density polyolefin obtained by polymerization making use of a metallocene catalyst, at least one alicyclic polymer layer and at least one thermoplastic resin layer.

5 [0007]

[Mode for Carrying out the Invention]

The film according to the present invention have a structure that at least one layer containing a linear low density polyolefin, at least one alicyclic polymer layer
10 and at least one thermoplastic resin layer are laminated.

[0008]

The linear low density polyolefin used in the present invention is a linear low density polyolefin having a long period of at most 275 angstroms as measured by a small
15 angle X-ray scattering method. The long period of the linear low density polyolefin is preferably at most 260 angstroms, more preferably at most 245 angstroms. When the linear low density polyolefin having a long period of at most 275 angstroms is used, the delamination of the
20 resulting laminated film becomes hard to occur and a laminated film excellent in mechanical strength can be provided.

[0009]

The lamellar thickness of the linear low density
25 polyolefin is preferably at most 145 angstroms, more preferably at most 140 angstroms. The resulting laminated film shows a tendency to more enhancing strength as the

lamellar thickness of the linear low density polyolefin becomes thinner.

[0010]

The linear low density polyolefin used in the present invention has a degree of crystallinity of usually 20 to 85%, preferably 35 to 70%, more preferably 45 to 65% as measured by large angle X-rays.

The density (a value measured in accordance with ASTM D 1505) of the linear low density polyolefin is usually 0.8 to 1 g/cm³, preferably 0.85 to 0.97 g/cm³. The melt flow rate of the linear low density polyolefin as measured at 190°C in accordance with ASTM D 1238 is usually 0.01 to 10 g/10 min, preferably 0.5 to 7 g/10 min. The melting point of the linear low density polyolefin is usually 80 to 150°C, preferably 90 to 130°C.

[0011]

No particular limitation is imposed on the production process of the linear low density polyolefin. However, those obtained by using a metallocene catalyst is preferred. As the metallocene catalyst, those conventionally used can be used. Examples of the metallocene catalyst include cyclopentadienyl derivatives of transition metals such as zirconium, titanium, hafnium, vanadium, niobium, tantalum and chromium, and combinations of such a cyclopentadienyl derivative and an organoaluminum compound such as aluminoxane.

The polymerization of the linear low density

polyolefin is preferably conducted by a vapor-phase process making no use of any solvent. The polymerization by the vapor-phase process tends to provide a linear low density polyolefin containing no unnecessary by-product. The
5 linear low density polyolefin obtained by the polymerization using the metallocene catalyst is used, thereby enhancing the mechanical strength and the like of the resulting laminated film.

[0012]

10 No particular limitation is imposed on the monomer composition of the linear low density polyolefin. For example, ethylene; and α -olefins such as propylene, 1-butene, 1-pentene, 1-hexene, 1-octene and 4-methyl-1-pentene may be used either singly or in any combination
15 thereof. Other copolymerizable monomers such as vinyl acetate may be used as copolymerizable components. In the present invention, a copolymer of ethylene and an α -olefin is preferable because the clouding of the resulting film is lessened, and delamination becomes hard to occur.

20 [0013]

The alicyclic polymer used in the present invention is a polymer having an alicyclic structure at its main chain and/or side chain. The alicyclic polymer preferably has the alicyclic structure at its main chain from the
25 viewpoints of mechanical strength, heat resistance and the like. Examples of the alicyclic structure include a cycloalkane structure, a cycloalkene structure and the like.

The cycloalkane structure is preferred from the viewpoints of mechanical strength, heat resistance and the like. Examples of the alicyclic structure include a monocycle, polycycle, a fused polycycle, a crosslinked cycle and
5 combined polycycles thereof. No particular limitation is imposed on the number of carbon atoms forming the alicyclic structure. However, it is within a range of generally 4 to 30 carbons, preferably 5 to 20 carbons, more preferably 5 to 15 carbons. Various properties such as mechanical
10 strength, heat resistance and moldability are balanced with one another at a high level by such an alicyclic structure.
[0014]

A proportion of the repeating unit having the alicyclic structure in the alicyclic structure polymer can
15 be suitably selected as necessary for the end application intended. However, it is generally at least 30% by weight, preferably at least 50% by weight, more preferably at least 70% by weight. The upper limit of the proportion is 100% by weight. If the proportion of the repeating unit having
20 the alicyclic structure in the alicyclic polymer is too low, the heat resistance of the alicyclic polymer becomes poor. It is hence not preferable to contain such a repeating unit in a too low proportion. No particular limitation is imposed on other repeating units than the repeating unit
25 having the alicyclic structure in the alicyclic polymer, and they are suitably selected as necessary for the end application intended. More specifically, as the alicyclic

polymer, may be used not only a homopolymer of a monomer having an alicyclic structure or a copolymer of such alicyclic monomers, but also a copolymer of the alicyclic monomer with a non-alicyclic monomer copolymerizable therewith. The resultant alicyclic polymer may be subjected to such a treatment that an unsaturated bond is hydrogenated into a saturated bond.

[0015]

As examples of the alicyclic polymer, may be mentioned norbornene polymers, monocyclic cycloolefin polymers, cyclic conjugated diene polymers, vinyl cyclic hydrocarbon polymers, and hydrogenated products thereof. Among these, the norbornene polymers and hydrogenated products thereof, and the cyclic conjugated diene polymers and hydrogenated products thereof are preferred.

[0016]

No particular limitation is imposed on the norbornene polymers, and examples thereof include polymers obtained by subjecting a norbornene monomer to polymerization in accordance with, for example, the process disclosed in Japanese Patent Application Laid-Open No. 14882/1991 or 122137/1991. Specific examples of the norbornene polymers include ring-opening polymers of norbornene monomer(s) and hydrogenated products thereof, addition polymers of norbornene monomer(s) and addition polymers of a norbornene monomer and a vinyl compound. Of these, the hydrogenated products of the ring-opening polymers of the norbornene

monomer(s), addition polymers of norbornene monomer(s) and addition polymers of a norbornene monomer and a vinyl compound copolymerizable therewith are particularly preferred from the viewpoints of good balance between heat resistance, dielectric constant, and the like.

[0017]

The norbornene monomers are publicly known monomers disclosed in the above-described publications, Japanese Patent Application Laid-Open Nos. 227424/1990 and 276842/1990, etc. Examples of the norbornene monomers include bicyclo[2.2.1]hept-2-ene (trivial name: norbornene), 5-methyl-bicyclo[2.2.1]hept-2-ene, 5,5-dimethyl-bicyclo[2.2.1]hept-2-ene, 5-ethyl-bicyclo[2.2.1]hept-2-ene, 5-butyl-bicyclo[2.2.1]hept-2-ene, 5-hexyl-bicyclo[2.2.1]hept-2-ene, 5-octyl-bicyclo[2.2.1]hept-2-ene, 5-octadecyl-bicyclo[2.2.1]hept-2-ene, 5-ethylidene-bicyclo[2.2.1]hept-2-ene, 5-methylidene-bicyclo[2.2.1]hept-2-ene, 5-vinyl-bicyclo[2.2.1]hept-2-ene,

[0018]

5-propenyl-bicyclo[2.2.1]hept-2-ene, 5-methoxycarbonyl-bicyclo[2.2.1]hept-2-ene, 5-cyanobicyclo[2.2.1]hept-2-ene, 5-methyl-5-methoxycarbonylbicyclo[2.2.1]hept-2-ene, 5-ethoxycarbonyl-bicyclo[2.2.1]hept-2-ene, bicyclo[2.2.1]hept-5-enyl 2-methylpropionate, bicyclo[2.2.1]hept-5-enyl 2-methyloctanate,

[0019]

Bicyclo[2.2.1]hept-2-ene-5,6-dicarboxylic acid

anhydride, 5-hydroxymethylbicyclo[2.2.1]hept-2-ene, 5,6-di(hydroxy- methyl)-bicyclo[2.2.1]hept-2-ene, 5-hydroxyisopropyl-bicyclo[2.2.1]hept-2-ene, 5,6-dicarboxy-bicyclo[2.2.1]hept-2-ene, bicyclo[2.2.1]hept-2-ene-5,6-dicarboxylic acid imide, 5-cyclopentyl-bicyclo[2.2.1]hept-2-ene, 5-cyclohexyl-bicyclo[2.2.1]hept-2-ene, 5-cyclohexenyl-bicyclo-[2.2.1]hept-2-ene, 5-phenyl-bicyclo[2.2.1]hept-2-ene,

[0020]

10 Tricyclo[4.3.0.1^{2,5}]dec-3,7-diene (trivial name: dicyclopentadiene), tricyclo[4.3.0.1^{2,5}]dec-3-ene, tricyclo[4.4.0.1^{2,5}]undec-3,7-diene, tricyclo[4.4.0.1^{2,5}]undec-3,8-diene, tricyclo[4.4.0.1^{2,5}]undec-3-ene, tetracyclo[7.4.0.1^{10,13}.0^{2,7}]tridec-2,4,6,11-tetraene
15 (another name: 1,4-methano-1,4,4a,9a-tetrahydrofluorene), tetracyclo[8.4.0.1^{11,14}.0^{3,8}]tetradec-3,5,7,12-11-tetraene (another name: 1,4-methano-1,4,4a,5,10,10a-hexahydro-anthracene),

[0021]

20 Tetracyclo[4.4.0.1^{2,5}.1^{7,10}]-dodec-3-ene (trivial name: tetracyclododecene), 8-methyltetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-ethyltetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-methylidenetetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-ethylidenetetracyclo-
25 [4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-vinylnitetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-propenyltetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-methoxycarboxy-tetracyclo-

[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-methyl-8-methoxycarbonyl-
 tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-hydroxymethyl-
 tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-carboxy-
 tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene,

5 [0022]

8-cyclopentyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene,
 8-cyclohexyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-
 cyclohexenyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-
 phenyl-tetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene,
 10 pentacyclo[6.5.1.1^{3,6}.0^{2,7}.0^{9,13}]-pentadec-3,10-diene and
 pentacyclo[7.4.0.1^{3,6}.1^{10,13}.0^{2,7}]-pentadec-4,11-diene.

[0023]

These norbornene monomers may be used either singly
 or in any combination thereof. The norbornene polymer may
 15 be a copolymer of a norbornene monomer and another monomer
 copolymerizable therewith. The content of bound norbornene
 monomer units in the norbornene polymer is suitably
 selected as necessary for the end application intended.
 However, it is generally at least 30% by weight, preferably
 20 at least 50% by weight, more preferably at least 70% by
 weight, whereby the dielectric constant, the heat
 resistance and elongation properties of such a polymer can
 be balanced with each other at a high level.

[0024]

25 Examples of the vinyl monomer copolymerizable with
 the norbornene monomer include ethylenes or α -olefins
 having 2 to 20 carbon atoms, such as ethylene, propylene,

1-butene, 1-pentene, 1-hexene, 3-methyl-1-butene, 3-methyl-
1-pentene, 3-ethyl-1-pentene, 4-methyl-1-pentene, 4-methyl-
1-hexene, 4,4-dimethyl-1-hexene, 4,4-dimethyl-1-pentene, 4-
ethyl-1-hexene, 3-ethyl-1-hexene, 1-octene, 1-decene, 1-
5 dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene and 1-
eicosene; cycloolefins such as cyclobutene, cyclopentene,
cyclohexene, 3,4-dimethylcyclopentene, 3-methylcyclohexene,
2-(2-methylbutyl)-1-cyclohexene, cyclooctene and 3a,5,6,7a-
tetrahydro-4,7-methano-1H-indene; and non-conjugated dienes
10 such as 1,4-hexadiene, 4-methyl-1,4-hexadiene, 5-methyl-
1,4-hexadiene and 1,7-octadiene. These vinyl monomers may
be used either singly or in any combination thereof.

[0025]

No particular limitation is imposed on the
15 polymerization process of the norbornene monomer, or the
norbornene monomer and the vinyl monomer copolymerizable
with the norbornene monomer, and a hydrogenation process.
The polymerization and hydrogenation may be conducted in
accordance with any publicly-known processes.

20 [0026]

As examples of the monocyclic cycloolefin polymer,
may be mentioned addition (co)polymers of a monocyclic
cycloolefin monomer such as cyclohexene, cycloheptene or
cyclooctene, which are disclosed in Japanese Patent
25 Application Laid-Open No. 66216/1989.

[0027]

As examples of the cyclic conjugated diene polymer,

may be mentioned (co)polymers obtained by subjecting a cyclic conjugated diene such as cyclopentadiene or cyclohexadiene to 1,2- or 1,4-addition polymerization, and hydrogenated products thereof, which are disclosed in Japanese Patent Application Laid-Open Nos. 136057/1994 and 258318/1995.

[0028]

As examples of the vinyl alicyclic hydrocarbon polymer, may be mentioned polymers of a vinyl cyclic hydrocarbon monomer such as vinylcyclohexene or vinylcyclohexane, and hydrogenated products thereof, which are disclosed in Japanese Patent Application Laid-Open No. 59989/1976, and polymers of a vinyl aromatic monomer such as styrene or α -methylstyrene, the aromatic ring portions of which have been hydrogenated, and which are disclosed in Japanese Patent Application Laid-Open Nos. 43910/1988 and 1706/1989.

[0029]

The alicyclic polymer may have a polar group such as a hydroxyl group or carboxyl group.

The alicyclic polymer having a polar group is obtained by, for example, (1) introducing a compound having a polar group into said alicyclic polymer by a modification reaction, or (2) copolymerizing a monomer containing a polar group as a comonomer.

Examples of the polar group include hydroxyl, carboxyl, oxy, epoxy, glycidyl, oxycarbonyl, carbonyloxy,

carbonyl and amino groups.

[0030]

The above-listed alicyclic polymers may be used either singly or in any combination thereof.

5 [0031]

No particular limitation is imposed on the molecular weight of the alicyclic polymer. The molecular weight of the alicyclic polymer is generally 1,000 to 1,000,000, preferably 5,000 to 500,000, more preferably 10,000 to 10 250,000 when expressed by a weight average molecular weight (Mw) in terms of polystyrene as measured by gel permeation chromatography (GPC) using cyclohexane or toluene as a solvent. It is preferable that the weight average molecular weight (Mw) of the alicyclic polymer falls within 15 this range because the heat resistance and adhesion property of the polymer and the surface smoothness of the resulting laminate are balanced with each other at a high level.

[0032]

20 The molecular weight distribution of the alicyclic polymer is expressed by a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the number average molecular weight (Mn) as measured by GPC using cyclohexane or toluene as a solvent and is generally at most 5, 25 preferably at most 4, more preferably at most 3.

The ranges of the weight average molecular weight (Mw) and molecular weight distribution (Mw/Mn) and the

measuring methods thereof are particularly suitable for the norbornene polymers. However, the present invention is not limited thereto. In the case of an alicyclic polymer the weight average molecular weight and molecular weight distribution of which cannot be measured by the above-described method, that having a melt viscosity or polymerization degree to such an extent that a resin layer can be formed by an ordinary melt processing process may be used.

10 [0033]

The glass transition temperature of the alicyclic polymer may be suitably selected as necessary for the end application intended. However, it is generally at least 50°C, preferably at least 70°C, more preferably at least 100°C, most preferably at least 125°C.

[0034]

Examples of the thermoplastic resins include polyolefins other than the above-described linear low density polyolefins such as polyethylene, polypropylene, polybutene and polypentene; polyester such as polyethylene terephthalate and polybutylene terephthalate; polyamide such as nylon 6 and nylon 66; ethylene-ethyl acrylate copolymers, ethylene-vinyl acetate copolymers and polycarbonate. Among these, polyethylene and polypropylene other than the above-described linear low density polyolefins are preferred.

[0035]

In the present invention, various kinds of components such as additives may be incorporated into the above-described linear low density polyolefins, the alicyclic polymers or the thermoplastic resins as needed. No particular limitation is imposed on the additives so far as they are those generally used in the field of resin industry. Examples thereof include hardening agents, hardening accelerators, hardening aids, fillers, heat stabilizers, weather stabilizers, flame retardants, leveling agents, antistatic agents, slip agents, anti-blocking agents, anti-clouding agents, lubricants, dyes, pigments, natural oils, synthetic oils, waxes, antioxidants, ultraviolet absorbents, light stabilizers, colorants, etc. These additives may be used in a proper amount within limits not impeding the objects of the present invention.

[0036]

In the present invention, a rubbery polymer may be incorporated into the above-described linear low density polyolefins, the alicyclic polymers or the thermoplastic resins from the viewpoints of imparting strength or flexibility to the resulting films.

Examples of the rubbery polymer include diene rubbers such as natural rubber, polybutadiene rubber, polyisoprene rubber, acrylonitrile-butadiene copolymer rubber, styrene-butadiene copolymer rubber, styrene-isoprene copolymer rubber and styrene-butadiene-isoprene copolymer rubber; hydrogenated products of these diene rubbers; saturated

polyolefin rubbers, such as ethylene- α -olefin copolymers such as ethylene-propylene copolymers, and copolymers of propylene with another α -olefin; α -olefin-diene copolymer rubbers such as ethylene-propylene-diene copolymers, α -
5 olefin-diene copolymers, isobutylene-isoprene copolymers and isobutylene-diene copolymers; special rubbers such as urethane rubber, silicone rubber, polyether rubber, acrylic rubber and ethylene-acrylic rubber; thermoplastic elastomers, such as aromatic vinyl polymers such as
10 styrene-butadiene-styrene block copolymer rubber and styrene-isoprene-styrene block copolymer rubber, and hydrogenated products thereof; urethane-based thermoplastic elastomers; polyamide-based thermoplastic elastomers; and 1,2-polybutadiene-based thermoplastic elastomers.

15 [0037]

The rubbery polymers may be used either singly or in any combination thereof. The blending proportion of the rubbery polymer is generally at most 100 parts by weight, preferably at most 70 parts by weight, more preferably at
20 most 50 parts by weight per 100 parts by weight of the above-described linear low density polyolefin, the alicyclic polymer or thermoplastic resin, and the lower limit thereof is 0 part by weight.

[0038]

25 The film according to the present invention may be any laminate so far as it is a laminate obtained by laminating at least one alicyclic polymer layer, at least

one layer containing the above-described linear low density polyolefin, and at least one thermoplastic resin layer. These layers may be each provided as either a singly layer or plural layers.

5 Specific examples of the laminate according to the present invention include a laminate obtained by laminating the thermoplastic resin layer, the layer containing the above-described linear low density polyolefin, and the alicyclic polymer layer in this order; and a laminate
10 obtained by laminating the thermoplastic resin layer, the layer containing the above-described linear low density polyolefin, the alicyclic polymer layer, the layer containing the above-described linear low density polyolefin, and the thermoplastic resin layer in this order.
15 The layer containing the above-described linear low density polyolefin is interposed between the thermoplastic resin layer and the alicyclic polymer layer, whereby the film which is hard to cause delamination and excellent in mechanical strength can be obtained.

20 [0039]

 The layer containing the above-described linear low density polyolefin may be any layers so far as it contains the above-described linear low density polyolefin. For instance, it includes a layer composed of the above-
25 described linear low density polyolefin alone, a layer of a composition containing the above-described linear low density polyolefin and another thermoplastic resin, a layer

of a composition containing the above-described linear low density polyolefin and an alicyclic polymer, a layer of a composition containing the above-described linear low density polyolefin, an alicyclic polymer and another thermoplastic resin. Of these, preferably used are the layer composed of the above-described linear low density polyolefin alone and the layer of the composition containing the above-described linear low density polyolefin and the alicyclic polymer.

10 [0040]

In the composition containing the above-described linear low density polyolefin and the alicyclic polymer according to the present invention, no particular limitation is imposed on the proportion of each component. However, from the viewpoint of preventing clouding, retaining high transparency and preventing delamination, a proportion [weight ratio] of the linear low density polyolefin/the alicyclic polymer is preferably brought into the range of generally 10/90 to 99/1, preferably 30/70 to 95/5, more preferably 40/60 to 90/10.

[0041]

The thickness of the film according to the present invention may be suitably selected as necessary for the end application intended. The thickness of the film is generally 0.5 μm to 5 mm, preferably 1 μm to 2 mm, more preferably 5 μm to 1 mm.

In the film according to the present invention, no

particular limitation is imposed on the thickness of each of the thermoplastic resin layer, the layer containing the above-described linear low density polyolefin and the alicyclic polymer layer. However, from the viewpoint of enhancing transparency or reducing clouding, the thickness of the thermoplastic resin layer is generally 0.09 to 1,000 μm , preferably 0.15 to 500 μm , the thickness of the alicyclic polymer layer is generally 0.3 to 2,000 μm , preferably 0.65 to 600 μm , and the thickness of the layer containing the above-described linear low density polyolefin is generally 0.02 to 1,000 μm , preferably 0.05 to 400 μm .

[0042]

The laminates according to the present invention can be provided by forming the respective layers by a melt forming method or solution casting method and then laminating the layers on each other. The laminates can also be obtained by melt forming the polymer or the resin making up the respective layers.

As a specific example of the solution casting method, may be mentioned a method in which a bar coater, T-die coater, T-die equipped with a bar, doctor knife, Meyer bar, roll coater, die coater or the like is used. A method of coating by means spraying, brushing, rolling, spin coating, dipping or the like may also be adopted. When the desired coating thickness cannot be achieved by one coating, the coating may be conducted repeatedly.

Specific examples of the melt forming method include melt-extruding methods such as a method making use of a T-die and an inflation method, a calendering method, a hot pressing method, and an injection molding method. Of these, melt-extruding methods are preferable.

[0043]

The extrusion conditions in the melt extrusion are the same as general conditions used for resin materials having almost the same glass transition temperature as the above-described resins. For example, the resin materials are extruded at a die temperature of about 230 to 300°C, and the extruded film is taken up by take-up rolls preset to a temperature of about 30 to 150°C to cool the film slowly or rapidly. In order to lessen surface defects such as die lines, it is preferable to construct an extruder in such a manner that portions at which the resin remains are lessened as much as possible. Therefore, it is preferable to use a die free of flaw and the like within the interior or lip thereof as much as possible.

[0044]

In order to laminate the respective layers, the resin materials for forming the respective layers may be co-extruded. From the viewpoint of dimensional stability, however, it is preferable to separately extrude the resin materials for forming the respective layers and then laminate the respective layers by calendering, rolling or the like.

[0045]

[EXAMPLES]

The film according to the present invention and the production process thereof will hereinafter be described more specifically by the following Examples. Evaluations were conducted in accordance with the following respective methods:

(1) Haze was measured in accordance with ASTM D 1033.

(2) Appearance upon stretching:

After stretching a laminate sample, the appearance of the sample was observed to evaluate whether clouding, wrinkling and torn line occurred or not.

(3) Delamination test:

After stretching a laminate sample, the separated state of the respective layers was visually observed.

[0046]

[Comparative Example 1]

A ring-opening copolymer of tricyclo[4.3.0.1^{2,5}]dec-3,7-diene with 8-ethyltetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene was hydrogenated, thereby obtaining a hydrogenated ring-opening polymer having a weight average molecular weight of 36,000, a molecular weight distribution (Mw/Mn) of 2.1 and a glass transition temperature of 136°C.

Thirty parts by volume of this hydrogenated ring-opening polymer were extruded by an extruder to form a hydrogenated ring-opening polymer layer. On the other hand, 30% by volume of polypropylene [product of Nippon

Polyolefin K.K., SG510, Vicat softening point: 135°C, density: 0.9 g/cm³, melt flow rate: 0.65 g/10 min., melting point: 148°C (DSC method)] were extruded by an extruder to form a polypropylene layer. The hydrogenated ring-opening polymer layer and polypropylene layer were then rolled by calender rolls to laminate them on each other, thereby producing a laminated film having a thickness ratio of the polypropylene layer/the hydrogenated ring-opening polymer layer of 1/1 and a total thickness of about 260 μm. The haze of this laminated film was 17. When stress was applied to this laminated film by stretching and folding it, clouding, wrinkling and/or torn line occurred. The polypropylene layer and the hydrogenated ring-opening polymer layer underwent delamination, whereby the respective layers could be separated from each other.

[0047]

[Example 1]

As linear low density polyethylene, was used linear low density polyethylene having a lamellar thickness of 130 angstroms, a long period of 235 angstroms, a degree of crystallinity as measured by X-rays of 54%, a density of 0.93 g/cm³, a melt flow rate of 1.7 g/10 min., a Vicat softening point of 110°C and a melting point of 121°C. This linear low density polyethylene is an ethylene-hexene copolymer obtained by polymerization making use of a metallocene catalyst obtained by adding N,N-dimethylanilinium tetrakis(pentafluorenyl) borate to a

mixture of diphenylmethylen- (cyclopentadienyl) (fluorenyl) zirconium dichloride and triisobutylaluminum. Six parts by volume of this linear low density polyethylene were extruded by an extruder to form a linear low density polyolefin layer, 30 parts by volume of the hydrogenated ring-opening polymer were extruded by an extruder to form a hydrogenated ring-opening polymer layer, 30 parts by volume of polypropylene (SG-510) were extruded by an extruder to form a polypropylene layer, and the linear low density polyolefin layer was held between the polypropylene layer and the hydrogenated ring-opening polymer layer to bring them into close contact with one another. The respective layers were rolled by calender rolls to laminate them, thereby obtaining a laminated film laminated in order of the polypropylene/the linear low density polyolefin/hydrogenated ring-opening polymer and having a thickness ratio of 10/2/10 and a total thickness of about 260 μm . This laminated film had the same haze value, 17 as the laminated film of Comparative Example 1 though it had a 3-layer structure. When stress was applied to this laminated film by stretching and folding it, none of clouding, wrinkling and torn line occurred. Further, the polypropylene layer and the hydrogenated ring-opening polymer layer were completely bonded through the linear low density polyolefin layer, and so no delamination occurred. In addition, this laminated film was also excellent in tearability and cuttability.

[0048]

[Example 2]

Seven parts by weight of the above-described linear low density polyolefin and 3 parts by weight of the above-described hydrogenated ring-opening polymer were kneaded, and 6 parts by volume of the kneaded product was extruded by an extruder to form a layer containing the linear low density polyolefin and the hydrogenated ring-opening polymer. A laminated film having a total thickness of about 260 μm was then obtained in the same manner as in Example 1 except that this layer was replaced for the linear low density polyolefin layer in Example 1. The laminated film thus obtained had a haze of 20 though the layer containing the hydrogenated ring-opening polymer and the linear low density polyolefin was laminated. When stress was applied to this laminated film by stretching and folding it, none of clouding, wrinkling and torn line occurred. Further, the respective layers were fully bonded to one another, and so no delamination occurred. In addition, this laminated film was also excellent in tearability and cuttability.

[0049]

[Effects of the Invention]

The films according to the present invention are free of clouding, high in transparency and heat resistance, excellent in tearability or cuttability, low in moisture permeability, hard to cause delamination, and good in

adhesion to others, and thus can be used in a wide variety of use applications such as food packaging films, drug packaging films and agricultural films.

[Document Name] ABSTRACT

[Abstract]

[Object] To provide a film which is free of clouding, high in transparency and heat resistance, excellent in
5 tearability or cuttability, low in moisture permeability and good in adhesion to others, and a production process thereof.

[Constitution] To provide a laminated film wherein a layer containing a linear low density polyolefin which has a long
10 period of at most 275 angstroms as measured by the small angle X-ray scattering method, preferably which is obtained by polymerization making use of a metallocene catalyst and further preferably which has a lamellar thickness of at
most 145 angstroms, is interposed, as an adhesive layer,
15 between an alicyclic polymer layer and a thermoplastic resin layer.

[Selected Figure of Drawings] None